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Framework for the Life Cycle Assessment of non-permanent process units in volatile chemical recycling process chains

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Abstract

To handle the rising amount of plastic waste in the environment, new technologies are necessary to recover the value of this resource. Chemical recycling is one option to treat mixed and multi-layer plastic waste streams. Emerging technologies works e.g. with pyrolysis (via thermal or catalytic cracking) of the plastic waste streams and results in multi-material output streams. Membranes are a widely adopted and a relevant technology for bulk separations, within chemical recycling operations. Based on a different separation principle, they need less thermal energy than e.g. distillation columns. However, when it comes to their contribution to the environmental impact of the whole process chain, membranes need special attention. Compared to installed process units, such as pyrolysis and catalytic upgrading reactors, membranes have a shortened life time (non-permanent) compared to the whole process chain and need extra treatments to avoid fouling and maintain their efficiency. These points make it challenging to integrate a LCA model representing membranes within the whole LCA process model. In this paper a framework for the LCA of membranes will be presented regarding their impact in a chemical recycling process and how a shortened life time model can be integrated into a longer life LCA process model with focus on the handling of the different time scales.

Keywords: chemical plastic recycling, membrane separation, LCA

1. Introduction

Plastics became one of the most important materials in our society. Due to their cost effectiveness and properties they are versatile and can be found in medicine, electronics, mobility, construction and the packaging industry [1]. Nevertheless, the high use of plastic products with a short life time such as packaging is the reason for the increasing amount of plastic waste [2]. Plastics can have significant environmental issues. Especially in the production and end-of-life phase of a plastic containing material, environmental issues occur, like the, use of the fossil fuels, pollution of the oceans with effects on the marine life or flooding countries without robust waste management systems. [3][4]. Especially, due to lacking waste collection systems in developing countries and inefficient recycling technologies, more and more plastic waste enter and damage the environment [4]. Current most common way to preserve plastic as a resource in the closed loop, is the mechanical recycling, which involve sorting by plastic type, melting and reprocessing them into a reusable resource per plastic type [3]. However, due the high requirements of clean, high-purity mono-material input streams, approximately 30 % of the annual European plastic waste can be treated using mechanical recycling. Consequently, the remaining 70 % of plastic waste is not recycled due economic or technical reasons [5]–[7].

Other end-of-life strategies such as incineration and landfill cannot completely close the gap in the circular economy, as

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either the resource or energy potential is no longer available [8]. To tackle this challenge, new recycling technologies are being developed to increase the recycling rates and to foster a closed loop economy.

Chemical recycling is in this regard seen as an alternative to conventional mechanical material recycling routes. By cracking up the plastic waste using chemical reactions, new added-value chemicals can be produced [3]. One possibility is the cracking of plastic waste by the use of pyrolysis. In this process, polymer compounds are broken down into high-quality chemical fraction, which are treated towards added-value chemicals with the use of various separation and downstream processes. One technical advantage of this technology, in comparison to mechanical recycling alternatives, is that the plastic waste stream does not need to be sorted by type and does not require expensive cleaning before the processing [9]. This means that even plastic waste fractions that are not suitable for mechanical recycling can be treated.

Nevertheless, chemical recycling also faces environmental challenges. Current studies show that the disadvantages of chemical recycling currently dominate the advantages. In particular, processes such as pyrolysis and gasification release environmental toxins such as bisphenol-A, benzene and volatile organic components [10], [11].

In order for chemical recycling technologies to be successfully established as a commercial alternative to current plastic recycling processes, it needs to be both environmental and economic advantageous. Decisions which are made in the early beginning of the process development have a far-reaching influence on the future functionality, costs and environmental impact [12]. For this reason, the further development of the technology should be accompanied by a life cycle assessment and life cycle costing at an early stage of development. Due to the non-pure waste streams, there is a high volatility within the process, since the splitting and treatment of the different polymer structures requires different amounts of energy.

Due to the constantly changing operating conditions, a static LCA, according to the ISO 14040/44 [13], [14], cannot reflect the changes in the system. Furthermore, it is necessary to consider the fact that a process chain can be composed of a wide variety of unit processes characterized by having heterogeneous life times. A process unit with a shorter life time than the general process chain is here defined as a non-permanent unit process.

This paper presents a modelling approach, to assess the effect of non-permanent process units in a permanent process system during the whole technology development and the operation of the industrial process plant.

2. Relevance of non-permanent process units: Use of membranes within chemical recycling processes

Membranes are an example of devices with a reduced life-time, compared to the process plants in which they are installed Membranes usually have a life time in a process of approximately five to eight years, while the life time of a process plant range from 25 to 50 years [15] [16]. In addition, they require extensive cleaning and maintenance during operation. The approach to be developed must therefore not only be able to map the volatile process parameters and flows within the process, but also consider the different lifetimes of the individual process units. This is especially relevant in the development stage of a process, since different options and also technological progress (TRL) within a process unit can lead to changes in the model. Membranes are a widely adopted and a relevant technology for bulk separations, within chemical recycling operations. Based on a different separation principle, they need less thermal energy than e.g. distillation columns and are therefore interesting for an efficient process operation.

Membranes are selectively permeable layers that can be used for separation. The selectivity creates a permeate that passes through the membrane and a retentate which remains on the upstream side of the membrane [17].

Previous studies have shown that the environmental effects caused by membranes are mainly generated during operation and disposal. Membrane production contributes only less than 5% to the environmental impact.[9],[10] Special challenges occur when assessing the operation of membranes. Besides a high energy demand during bulk separation, membranes require complex cleaning after each use to avoid fouling on the separation layers. Harmful and highly flammable solvents are often used for this purpose. These facts show that process units need special attention when it comes to the life cycle assessment.

In Table 1 the current state of research is shown to derive the research gap. For a comprehensive review four categories are considered.

Table 1: State of research-LCA of membranes in chemical recycling

Categories / References	Chemical recycling of plastic waste	Process industry	Membranes in chemical recycling	Waste water treatment & desalination
Rickert et al. [20]	●	●	○	○
Russ et al. [10]	●	●	○	○
Rollinson et al. [11]	●	●	○	○
Lawler et al. [16]	○	●	○	
Vince et al. [21]	○	●	○	●
Hancock et al. [18]	○	●	○	●

● considered ○ not considered

All references considered are related to LCA in the process industry. However, they examine either chemical recycling or the use of membranes in wastewater treatment or desalination. In chemical recycling, either the entire process or the main process is considered. Process steps for cleaning the membranes are not considered separately.

Membranes have been playing an important role in wastewater treatment and desalination for quite some time. The current state of research shows that the LCA of non-permanent process units in chemical recycling is not considered yet. It supports the approach to develop a dynamic framework for the assessment of non-permanent process units in chemical recycling with the use of live data from the plant to avoid unwanted process states.

3. Approaches for the consideration of volatile processes in LCE

It is necessary to further develop the static framework towards a dynamic LCA framework, which is able to consider the process changes or states in a given time frame. This is particularly important in chemical recycling, as the process must prove to be advantageous over other processes. It needs to be more advantageous than other recycling technologies, as well as the conventional production processes of the end products, in both environmental and economic terms. If the dynamic LCA approach makes this possible, the stakeholder is able to define necessary criteria to remain competitive. Due to the changing input streams, preliminary studies of operating conditions have shown that not every composition of the input stream is environmental and economically feasible to recycle, unless its negative impact can be compensated by comparatively positive compositions. Therefore, the approach must be suitable for production planning as well.

In most chemical industrial plants, a comprehensive process control system is installed, in which all data strings converge. This fact can be used for the development of a new LCA system architecture, because apart from the LCA model, only a data transfer in real time has to be integrated into the process control in order to transfer the necessary data into the Life Cycle Inventory. A first approach, how such a system could look like were given by *Hagen et al.* [22]. Here a dynamic LCA system was developed, which made it possible to send live data from a pilot factory to an LCA model. Data is collected in a high temporal resolution and the resulting environmental effects are immediately calculated. The approach supports dynamic decision making within process development processes. Differences in the process, caused by volatile input parameters become transparent. The results show that the potential of live data to assess volatile process systems is enormous.

4. Concept for the consideration of non-permanent unit processes in the LCA of Chemical Recycling processes

Analysis of requirements

Goal and scope

The goal is to assess non-permanent process units over the time in all TRL stages, from two perspectives. The first perspective is the comparison to recycling routes, the second perspective is the comparison to the conventional production processes. Due the fact that chemical recycling technologies always has to be advantageous in two scenarios, the developed system model has to consider both: chemical recycling as a recycling process and as a production process.

This leads to the requirement that the underlying model has to be flexible to switch easily the investigated scenario, without the need for two different models. As a reference process, in this case refer the iCAREPLAST process chain [9], which is a pilot plant for pyrolysis based plastic recycling, located in Zaragoza, Spain. This technology is currently at a TRL stage of 4-5 and should be at TRL 7 after the end of the project. Because of the changing TRL, the developed framework should consider multiple technology readiness levels. The concept should be able to support the stakeholder at each TRL in the decision making and on-going development of the process unit. The information about the life time should be in comparison with the expected life of the whole process chain, to estimate how much of the environmental impacts and costs can be related to one production batch.

As a non-permanent process unit three membranes with various operations tasks are included in the process chain. While some of the process units are already existing from previous pilot processes, the membranes are not included yet and need to be designed especially for this process. If the assumed life time of the investigated process is ten years, for a pilot plant and 25 years for an industry plant, the life time of membranes is with a life time of eight years shorter.

This motivates to find a solution, how these process units can be assessed in the process chain with keeping the uncertainty at the lowest degree possible and to support the stake holders in their decision making.

Functional unit and system boundary

Because the investigated system is assessed from two points of view, two functional units are needed. On the one hand the system is compared to the mechanical recycling. The functional unit has to be input-based to compare the two systems regarding their treatment of a specific amount of plastic waste. E.g. treatment of 100kg plastic waste per hour. On the other hand, the investigated system needs a functional unit to be compared against other production process for alkylate chemicals. E.g. production of 1kg alkylates.

This output-bases functional unit compare the processes regarding their amount of producing a specific amount of the wanted product. The processes which are directly linked to the process chain are embedded in a foregroundsystem, all processes which are not directly linked are embedded in the background system.

Structure of the Life Cycle Inventory for non-permanent process units

In the following section a structure for the life cycle inventory (LCI) for non-permanent process units is presented. The LCI data has to be structured in a way that future changes of the process unit (e.g. development towards a higher TRL) can be integrated.

A standardized structure for the inventory helps to better organize and, if necessary, exchange data and interfaces. The inventory is primarily structured according to the life cycle stages (production, operation, end-of-life) of a, here: membrane as a non-permanent process unit. The subdivision of the stages helps to introduce clear demarcations between the phases, as well as to differentiate between static and dynamic

data backups.
For stages such as the production stage of membranes, simulation models as well as manufacturer information can be stored without accessing the process control system. In the manufacturing stage data can be recorded directly from the process control system. If the membrane type is replaced due to technological progress, new manufacturer information can be stored without affecting the stored interfaces in the operating stage.

Raw material and production

As mentioned before the raw material and production stage is responsible for less than 5 % of the occurring environmental impact of a membrane. This amount is decreasing when considering the whole production chain. For this stage, depending on the TRL, a simulation model or manufacturer information can be used. Simulation models, as seen in Figure 1 can be linked into the LCA system. If any changes occur during the process due new data etc., these could be recognized in time.

this stage all inputs and output streams, auxiliaries and energy demands, used by the membrane, are considered.

An efficient operation of a membrane requires a high amount of maintenance activities, like the cleaning of the membrane after every use with solvents like toluene to reduce fouling. This procedure is named cleaning-in-place (CIP). As fouling is not possible to be completely avoided, it is necessary to deconstruct membranes and perform a more extensive cleaning (cleaning-out-place (COP)). This leads to production stops, which leads to downtime cost. Associated data can be measured directly in the plant live data, when it comes to the CIP cleaning. The data for the COP needs to be transmitted manually.

End-of-life

The common used end-of-life strategies for membranes are the disposal and incineration. Due their complex construction recycling is not yet economical feasible. There are already studies, where the different end-of-life strategies are investigated [16],[21]. Several studies look at reverse-osmosis membranes. The membranes show similarities to membranes used in chemical recycling with regard to the used materials. Thus the available LCI data can be used for the study.

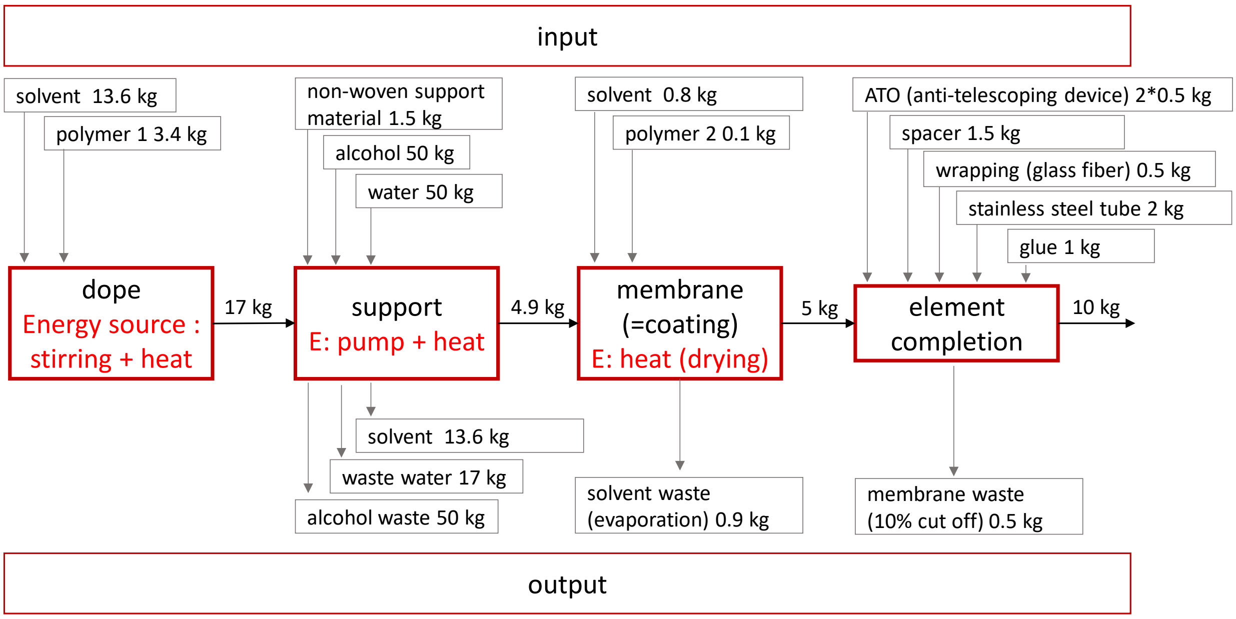

Figure 1: Simulation model for membrane production

Operation and maintenance

During operation of the membrane, the main environmental impacts occur. Depending on the composition of the used input stream the efficiency and the used resources by the membrane vary. To consider all changes, live data from the plant is used. This is possible from TRL 5 (Laboratory scale system validation in relevant environment) [12].
At lower TRL, there are typically no live data available and the data collection and processing needs to be done manually. In

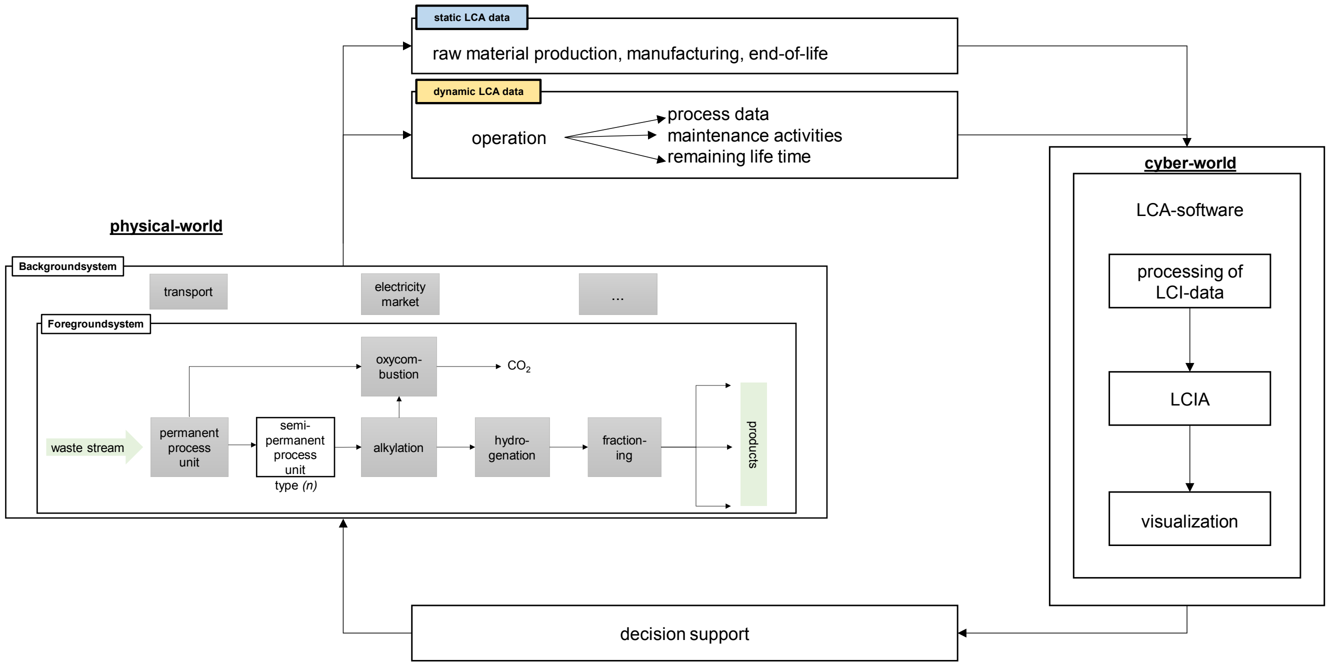

Figure 2: CPPS-framework for a dynamic consideration of volatile systems in LCE

5. Implementation concept

Based on the previously defined requirements for an assessment of non-permanent process units in volatile chemical recycling chains, the framework shown in Figure 2 was developed. The basis here is a cyber-physical production system (CPPS). It consists of a physical and a cyber -world. In the physical world, the real processes are integrated, such as process units, software and hardware, as well as the input and output streams. In the iCAREPLAST process, these are the plastic waste as input stream and several product output streams. The input stream is fed into the pyrolysis and the multi-fraction intermediate stream is broken down into the individual fractions by using downstream processes Here, the processes are divided in permanent and non-permanent process unit, depending on their life time in the process. The physical world is connected to the cyber world via various interfaces such as sensors etc. and transmit process data for performing an LCA. Each process from the physical world has a cyber-representative in the cyber world to evaluate their environmental impacts. They are organized in a foreground and a background system. In the foreground system are all process units integrated, which are directly linked to the chemical recycling process, while the background system, processes are integrated which are indirect linked to the recycling process. The individual process units are designed as blocks. All information and process sequences are stored in the respective process block in order to receive, process and forward the data of the respective processes from the physical world. The interlinkages between the individual process blocks are defined in a second logic layer in order to keep complexity and the data transfer rate at a low level. This enable to exchange the individual blocks without risk of affecting the structure.

This enables to assess different options of a selected process unit in the same structure without extensive changes within the model. The needed data is stored in a dynamic database, which is linked to the concerned process unit and contains all necessary data from each life cycle phase, as well as information about the TRL and the expected life time of the process unit technology. It's important to document the TRL of the investigated process unit in the database, because only on higher TRLs it's possible to receive data directly from the process in the physical world. On lower TRL stages data from the lab or from simulation models will be used.

The difference in the live and static data is mainly based in the fact, if there is data available, which can be received in real time or not and if its influence the environmental impact. If the influence on the environmental impact is low, static data is used to keep the error-proneness low. The static and live data are compiled into a data set and transferred to an LCA software via live links. The software calculates the LCA results and transfers them back to the database, where they can be used for decision support, such as driving the optimal operating point based on defined criteria. This can be done automatically or manually by the process operator. By means of suitable visualizations, different states and options can be compared with each other. This method is particularly suitable for processes under development as well as for non-permanent process units which, when replaced, have also frequently undergone parallel technological progress. Due to the block design, the new unit or technology can easily be integrated directly after completion without endangering the running operation or the stored model.

6. Discussion

In the following section the developed framework for the Life Cycle Assessment of non-permanent process units is discussed. Currently, the developed concept is not easily realizable in commercially available LCA software on the market. This hinders process operators and developers to use it for their processes. Due to the lack of a software solution, it is currently not possible to apply this modular system to other process systems without difficulty. Due to the high complexity of the process chain, it is not possible to develop modules for the flexible assessment of process units without further effort. The dependencies and interdependencies must be considered in the modelling. It must also be taken into account that as the level of technology advances, dependencies within the process chain may shift or changes may occur at other levels. Furthermore, it is to be expected that the non-permanent process units, depending on their nature, have only a minor influence on the overall balance.

However, the frame is able to combine different lifetimes and technology levels. By establishing this approach in the development process, it is possible for developers to combine different process combinations without having to adapt the process chain in the plant. Thus it may be possible to identify the most ecologically valuable option in advance.

7. Conclusion and Outlook

The necessity to accompany and support the development of new chemical recycling processes with a Life Cycle Assessment at an early stage is given by the currently still predominant environmental disadvantages. The goal was to develop a framework that allows to enter at the process level and combine different lifetimes and technology levels in order to support the process developers specifically at the respective point. The developed concept could already be tested in the iCAREPLAST process and showed good results there, within combining different process scenarios and testing process options easily. However, due to the complexity of the processes, interrelationships at other process levels must be more clearly recognizable to the user. For commercial use, software solutions must be created in the coming years.

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